Influences of inert nitrogen molecules, nitrogen radical atoms and nitrogen molecular ions on growth process and crystal structure of GaN heteroepitaxial layers grown on Si(001) and Si(111) substrates by molecular-beam epitaxy assisted by electron cyclotron resonance

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We have investigated the influences of inert nitrogen molecules, nitrogen radical atoms and nitrogen molecular ions on optical (photoluminescence) and crystallographic (X-ray diffraction and co-axial impact ion scattering spectroscopy) properties of GaN heteroepitaxial layers (heteroepilayers) grown on Si(001) and Si(111) substrates by molecular-beam epitaxy assisted by electron cyclotron resonance (ECR). GaN heteroepilayers were grown at 800℃, the temperature of Ga K-cell of 1041℃ and various nitrogen gas flow rates of 1.5-8 sccm by keeping the respective nitrogen plasma intensities of 391 and 357 nm-emission lines in ECR-plasma source at a μ-wave power of 350 W to about 100 and 320 arb.units, or by keeping the nitrogen plasma intensity density per 1sccm of the 357 nm-emission line to 160 arb.units/sccm under the identical 391 nm-emission line of 100 arb.units. The 357 nm- and 391 nm-emission lines are plasma emissions from nitrogen radical atoms and nitrogen molecular ions, respectively. The plasma intensity of the latter is maintained to a low level of 100 arb.units in order to suppress ion damage [1]. Figure 1 shows the growth rate dependence of nitrogen gas flow rate. The circles indicate a series of samples grown under a 357 nm-emission intensity of 320 arb.units and squares a series of samples grown under the identical 357 nm-emission intensity density per 1 sccm of 160 arb.units/sccm. The growth rates of the GaN heteroepilayers were remarkably decreased with an increase in the nitrogen gas flow rate when the nitrogen plasma emission intensity of 357 nm-emission line was constant. Therefore, the growth rate was deceased with a decrease in the nitrogen radical atoms. On the other hand, the growth rate was increased to the almost the same value even at a high nitrogen gas flow rate of 5 sccm by maintaining the same nitrogen plasma intensity density per 1 sccm of 357 nm-emission line as that of 2 sccm (160 arb.units/sccm) in ECR-plasma source. Thus, it indicates that the growth rate is dependent on effective V/III mole ratio rather than the nitrogen gas flow rate. This strongly suggests that the ECR-assisted growth process of GaN is governed by the effective V/III mole ratio, namely the ratio of density of the nitrogen radical atoms in inert nitrogen molecules to that of Ga atoms supplied from the Ga K cell. The inert nitrogen molecules in ECR-plasma obstruct the GaN growth and remarkably reduce the growth rate. The X-ray diffraction measurements show that the X-ray intensity of (333) diffraction

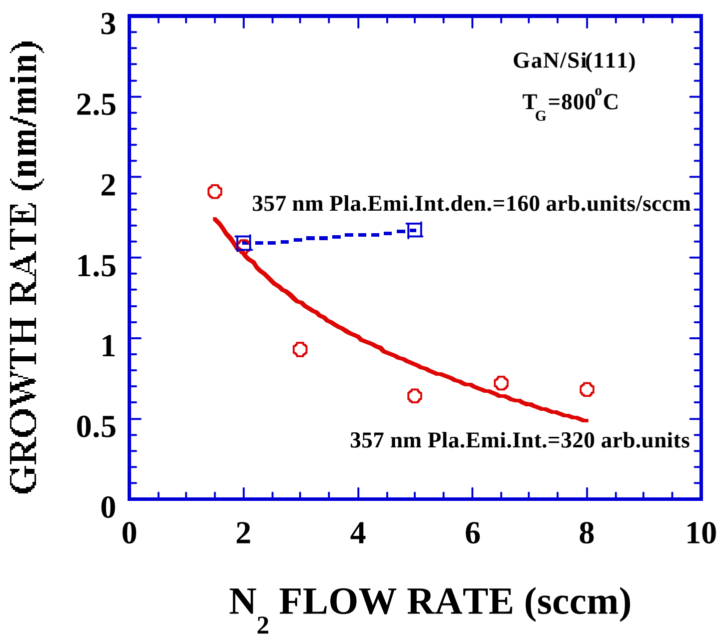

Fig. 1 Growth rate dependence of nitrogen gas flow rate

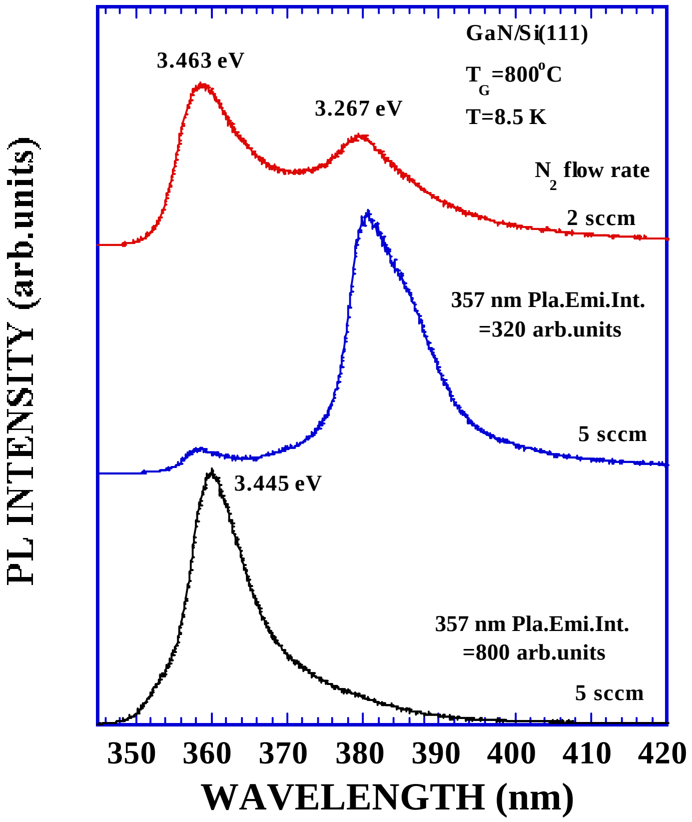

Fig. 2 8.5 K PL spectra near band-edge region of GaN heteroepilayers on Si(111) substrates

from cubic (β-) GaN grains was remarkably increased compared to that of (0006) diffraction from hexagonal (α-) GaN ones with an increase in the nitrogen gas flow rate when the nitrogen plasma emission intensity of 357 nm-emission line was constant. This reason is related to a change of the growth condition from N-rich condition to Ga-rich one, reflecting the decrease in the density of nitrogen radical atoms. Namely, the ratio of density of the nitrogen radical atoms in inert nitrogen molecules to that of Ga atoms reflects the effective V/III mole ratio in ECR-plasma process. The layer characteristics are governed by that. Figure 2 shows the 8.5 K PL spectra near the band-edge region of GaN heteroepilayers grown at a Ga K-cell temperature of 1041 ℃ and at two nitrogen gas flow rates of 2 and 5 sccm on Si(111) by keeping the respective nitrogen plasma intensities of 357 nm-emission lines to 320 and 800 arb.units, namely the different nitrogen plasma intensity densities per 1sccm of the 357 nm-emission line to 64 and 160 arb.units/sccm. These spectra clearly exhibited two exciton emissions from α- and β-GaN crystal at around 3.463 and 3.267 eV, respectively, which indicates that GaN heteroepilayers contain two kind of different crystal structures of α-GaN and β-GaN grains. The PL intensity of exciton emission from β-GaN was markedly increased compared to that from α-GaN with an increase in the nitrogen gas flow rate from 2 to 5 sccm when keeping nitrogen plasma intensity of 357 nm-emission lines to 320 arb.units, indicating the decrease in the ratio of density of the nitrogen radical atoms in inert nitrogen molecules to that of Ga atoms. It indicates the decrease in the effective V/III mole ratio, namely the change of the growth condition from N-rich condition into Ga-rich one. On the other hand, when the nitrogen plasma intensity of 357 nm-emission lines was increased from 320 to 800 arb.units at a nitrogen gas flow rate of 5 sccm, namely by keeping the identical nitrogen plasma intensity densities of the 357 nm-emission line to 160 arb.units/sccm, the PL spectrum dominantly exhibited exciton emission from α-GaN again. This clearly reflects the typical growth in N-rich condition [2]. It therefore indicates that the effective V/III mole ratio at 5 sccm is kept almost the same as that at 2sccm. It is concluded that the layer characteristics are governed by the effective V/III mole ratio in ECR-plasma process. Furthermore, it was experimentally confirmed that inert nitrogen molecules do not much degrade the optical and crystallographic properties of the grown GaN heteroepilayers although they obstruct the growth of GaN itself. We will also discuss the influences of inert nitrogen molecules, nitrogen radical atoms and nitrogen molecular ions on crystalline quality of GaN heteroepilayers on Si(001) substrates and on the growth mechanism of α-GaN and β-GaN in more detail in this conference.

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